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Liquid X-ray diffraction on the NIF: Phase transitions into and within the liquid phase

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NIF Facility Time Proposal:

Liquid X-Ray Diffraction on the NIF

Traditional Hugoniot velocity measurements have always been severely limited in their ability to determine structural properties of materials. However, the advent of x-ray diffraction¹ on laser compression experiments is recently allowing unprecedented opportunities to directly measure the in-situ structure of compressed materials. Unfortunately, much of the phase space accessible to shock compression in general and on the NIF specifically is in the liquid or fluid phases, so that traditional Bragg diffraction cannot be used. This proposal seeks beam time to address this issue by developing a reliable capability for liquid diffraction on the NIF.

There are currently three primary applications of liquid x-ray diffraction in high-pressure science. First, the substitution of liquid diffuse scattering for Bragg scattering is the preferred method for documenting melting in laser-heated DAC experiments.²⁻⁴ Second, the search for and observation of liquid-liquid phase transitions requires a direct structural diagnostic.⁵ And finally, great progress has been achieved in measuring static-high-pressure liquid diffraction in diamond-anvil cells (DACs) in the past decade.⁶⁻⁸

Scientific Discussion:

Melt transitions at ultra-high pressures and temperatures: Determining melt in shock experiments is extremely difficult. Flier plate experiments have used the release from the flier plate to measure the sound speed at high pressure.^{9,10} We have recently used abnormalities in the temperature of a decaying shock to determine the melt temperature.^{11,12} This is a powerful method, but is limited to transparent materials that become reflecting in response to the shock. It is our goal to develop sufficiently high quality liquid-diffraction data that a direct, positive identification of the liquid state can be made. As has been noted several times for DAC experiments lack of sharp diffraction is not sufficient to prove melt.^{2,4} Both iron^{4,9} and molybdenum^{3,10} have significant discrepancies between the melt determined in shock experiments (using the sound speed method, and various DAC techniques) that have not been resolved. It would be

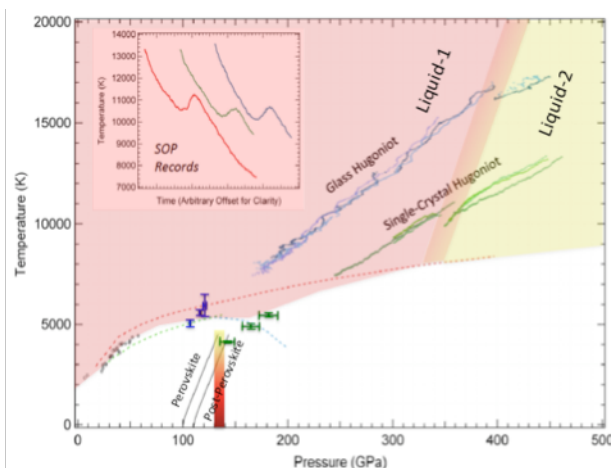


Fig. 1. Observations of dramatic changes in MgSiO₃ enstatite.

very useful to directly measure the shock melted liquid in a laser shock.

Liquid-liquid transitions at ultra-high pressures and temperatures: A long-standing debate about the existence of first order liquid-liquid structural changes was settled in 2000 by direct x-ray diffraction measurements on phosphorous.⁵ Probably the most important reason that liquid-liquid transitions are not more common is the difficulty in making accurate structural determinations. In phosphorous, the density change is 40%¹³, and the

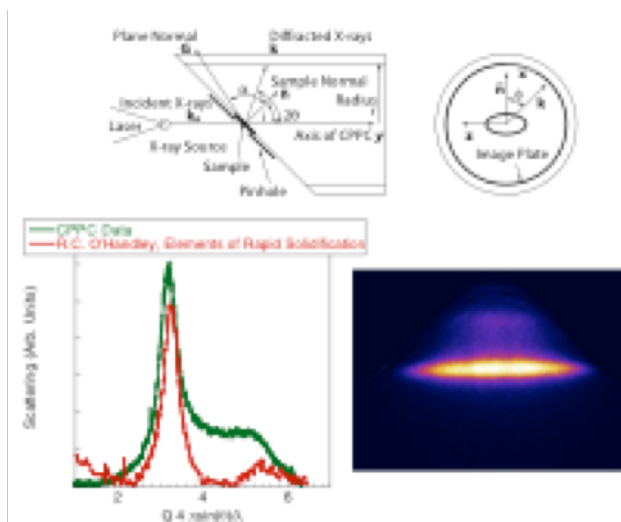


Fig. 2. Example data from single shot diffraction of amorphous MetGlas. A comparison is shown between the single shot data and published data using standard x-ray diffraction techniques.

transition is not subtle at all going from a tetrahedral-molecular to a network-atomic liquid structure.¹⁴ In order to see a signature of a liquid-liquid transition under laser compression, the structural change must be dramatic. Therefore we will concentrate on two materials with strong experimental evidence of dramatic change, enstatite and nitrogen. In enstatite we have performed numerous experiments similar to those used to determine the melt temperature of SiO₂ and diamond. We see very reproducible dramatic features at about 350GPa and 10000K that are difficult to include in a phase diagram without being a liquid-liquid transition (Fig 1). To confirm this interpretation, diffraction is essential. In nitrogen, anomalous single and double

shock data implied a continuous liquid-liquid transition at 30GPa and 6000K.¹⁵ A recent theoretical study confirms this interpretation and predicts a discontinuous transition at temperatures below 4000K.¹⁶

Methodology: There are three main technical issues with doing liquid diffraction on the NIF, a) sufficiently high scattering momentum, b) high background levels, c) the need for a reference background spectra. Single-shot diffraction from MetGlas on the Janus laser has shown that structure factors of amorphous materials can be measured on a nanosecond timescale in a single shot (Fig. 2). This shot was limited to a scattering momentum of $\sim 60 \text{ nm}^{-1}$ due to the Fe backlighter used (6.7 keV). Using higher Z backlighters like Mo (17.9 keV) or Ag (22.6 keV) would allow scattering momentums approaching 200 nm^{-1} as needed to obtain high quality structural information. The high background levels that we have generally found when doing very high pressure diffraction is caused by low energy x-radiation from the ablation plasma. Going to the high energy backlighters will also help to eliminate this background through the use of better low-energy filtering. Finally, the reference spectrum will have to use an undriven reference shot. It will be critical to make this reference as similar as possible to the actual shot and then use a corrective algorithm⁶ to account for differences.

A different experimental geometry, better suited for NIF is an energy dispersive diffraction using a broadband x-ray source and a single-photon counting CCD detector. We would use a capsule implosion target to generate the broad-band x-rays (different dopants can be used to optimize the signal for a particular material). With this geometry the detector can remain remote from the target, the detector can be electronically read out, and using a gated CCD we may be able to take both the reference and the background in the same shot cycle.

Experimental Feasibility:

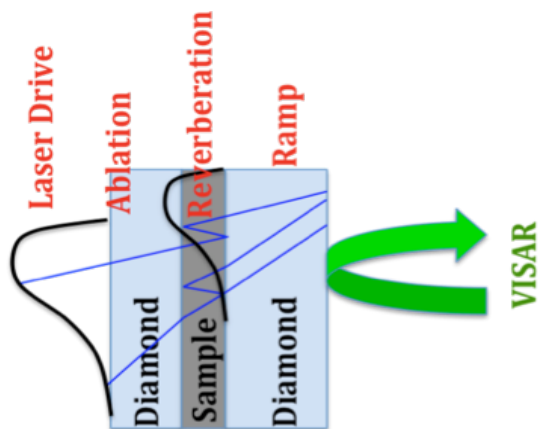


Fig. 3. Potential geometry and pulse-shape for initial optical measurements for ramp compressed diamond to 30 Mbar.

The experiments will be carried out on the 'materials science' NIF platform. Polycrystalline materials are interrogated with a collimated source quasi-monochromatic X-ray source. We use helium-alpha emission from a mid-Z element using a metallic x-ray source foil or foam. We will augment the diffraction data with VISAR, so that we obtain not only information about the crystal's phase and strain history, but also the stress profiles. The necessity for NIF is that we require the relatively long (several ns to tens of ns) drive, with its precision temporal shaping, in order to ultimately compress the material isentropically to pressures far above the normal point of melting on the shock Hugoniot. Our final goal is

to observe the phase and response of diamond at between 20 and 30 Mbar.

Methodology:

We propose to use our recent successes on Omega and Omega EP on diffraction of iron and diamond to 5 Mbar to design and perform our NIF experiments. The critical advance in these experiments is the use of a diamond-sandwich target. Much as a static diamond-anvil cell (DAC) sandwiches the sample between two strong diamond anvils, we sandwich a thin ($\sim 4 \mu\text{m}$) sample between two stiff diamond windows (Fig. 3). The first diamond acts as an ablator, transmitting a ramped-pressure drive generated by a ramped-intensity laser pulse. The second diamond acts as a tamper, maintaining a high pressure in the thin sample until reverberations from the free surface return to release the sample pressure. We probe the peak pressure state in the sample by carefully synchronizing a second laser pulse directed to an x-ray line-source target. Our primary pressure diagnostic is the free surface velocity history measured using a VISAR from the uncoated diamond-vacuum interface. The particle velocity-pressure calibration derived from other experiments³ directly yields the sample pressure. As long as the sample is sufficiently thin, it acts as a small perturbation to the diamond hydrodynamics and maintains a pressure in close agreement with the diamond windows. We have developed feasible NIF pulse shapes for diamond ramps to over 30 Mbar and conceptual drives to 100 Mbar. A slight perturbation of this geometry uses a

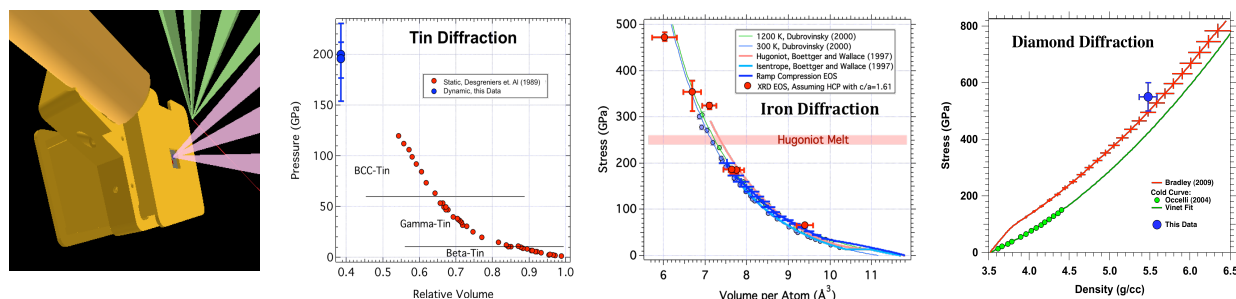


Fig. 4. X-ray diffraction configuration fielded at Omega. Unpublished diffraction results on solid tin, iron and diamond from Omega experiments.

LiF window in place of the second diamond. We have shown in experiments at Omega and Omega EP that LiF remains transparent to over 8 Mbar. This geometry would allow more direct examination of the sample pressure, synchronous optical measurements, and potential estimation of sample temperature.

At Omega and Omega EP we have successfully fielded a powder x-ray diffraction image plate (PXRDIP) holder 5 times (Fig. 4). We have measured diffraction on tin to 2 Mbar, on iron to 4.7 Mbar, and on diamond to over 5 Mbar. All three of these dynamic diffraction results significantly exceed the highest-pressure static experiments and represent the highest-pressure diffraction ever measured. The PXRDIP was designed to minimize the need for precise relative alignment of the x-ray source and the sample. The critical alignment needs are that the lasers hit the x-ray source, and the sample independently. This is done routinely at Omega and the NIF.

The NIF will allow us to significantly improve both the maximum pressure and precision of these diffraction experiments. We propose three different potential XRD diagnostics for NIF shown in Fig. 5. A) An image plate (IP) box very similar to the PXRDIP fielded at Omega. This geometry uses a halfraum drive geometry and a VISAR mirror mounted behind the back IP. All the unconverted light is incident on the horizontal front IP shield; in order for the x-ray source to avoid the unconverted light it must be placed directly in line with the sample. B) A pyramid-shaped IP box modeled after the broad-band x-ray diffraction (BBXRD) diagnostic fielded by members of this team on Omega. This geometry allows a NIF hohlraum drive with the unconverted light incident on the slanted sides of the IP box. The x-ray source can be placed at many angles with respect to the drive and still avoid the unconverted light. C) Finally, A Soller slit can shield the IP from unwanted x-radiation at the same time that the entire diagnostic is kept out of range of all unconverted light. This concept has not yet been tested on a laser platform, but has been shown to be very effective on high-pressure synchrotron beam lines.¹⁴ While the Soller slits eliminate the problems with unconverted light, an effective alignment procedure will need to be developed. All three proposed diagnostics involve relatively large targets or diagnostics placed close to TCC,

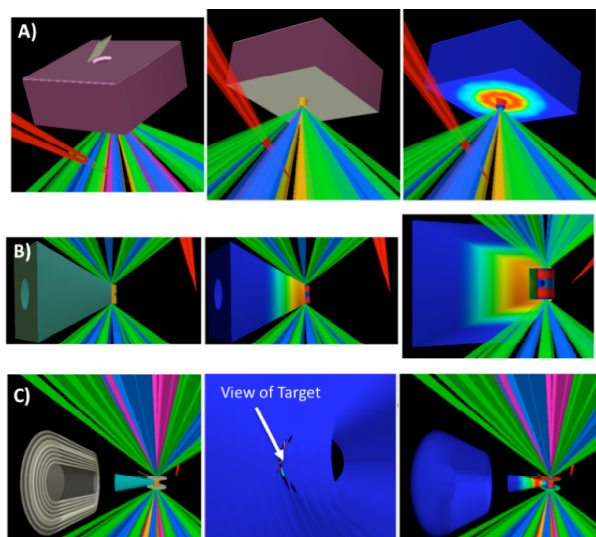


Fig. 5. Three possible configurations for an x-ray diffraction diagnostic on the NIF. A) IP box with hemispherical symmetry. Requires a 90° VISAR mirror, on-axis x-ray source, and detailed TALIS review of unconverted light, target alignment, and IP protection. B) IP pyramid, using the NIF's Hohlraum geometry, including an off-axis x-ray source. Requires a detailed TALIS review of unconverted light, target alignment, and IP protection. C) Single IP shielded by conical Soller slits, using the NIF's Hohlraum geometry, including an off-axis x-ray source, and avoiding all unconverted light. Requires a detailed TALIS review of diagnostic (Soller-slit) alignment.

and will require careful evaluation to be fielded at the NIF. We have begun to discuss these concepts with key members of the TALIS committee and are confident that by integrating NIF facility teams, a suitable XRD diagnostic package can be designed and fielded. There is also a great programmatic need for an XRD capacity on the NIF, and the substantial overlap between this team and the programmatic teams will ensure that an optimal diagnostic is fielded that is flexible enough to allow all desired XRD-based experiments to use a single diagnostic.

The diamond diffraction targets will be manufactured by Diamond Materials GmbH in Germany. The target box (with associated debris calculations) used for holding image plates is being developed in concert with the materials IET. We will also attempt to recover the sample using schemes under consideration and development by the materials IET.

Results Expected: We are currently developing all the methods proposed above on the Jupiter and Omega laser facilities. On the NIF we will begin to design and qualify the energy dispersive detector. Our first shots on NIF will be to try to detect a liquid-liquid transition in MgSiO_3 enstatite.

Desired Platform: The platform will be similar to the HED materials platform as described on the NIF experimental platform webpage.

Experimental Team:

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Dr. Gilbert Collins, LLNL, collins7@llnl.gov

In addition, this proposal is submitted in concert with all members of the Planetary Sciences University Use of NIF Consortium led by Raymond Jeanloz, UCB, jeanloz@berkeley.edu

Required Capabilities and Resources: Funding for LLNL scientists will come from existing LDRD's.

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- 7 Weck, G., Loubeyre, P., Eggert, J. H., Mezouar, M. & Hanfland, M. Melting line and fluid structure factor of oxygen up to 24 GPa. *Phys. Rev. B* **76**, 054121 (2007).

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Summary of proposed experiment (Page 1 of 3)



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- **_Desired platform (If known): *Materials***

(Available platforms include: a) Capsule implosions; b) Hohlraum energetics; c) Radiation transport; d) Shock timing; e) Streaked radiography; f) X-ray opacity; g) X-ray sources. For further information on platforms see the NIF website: https://lasers.llnl.gov/for_users/experimental_capabilities/index.php)

- **Number of shots requested: Please fill out table below indicating number of “good data” shots requested each year. Do not add in additional shots to account for contingency, experimental problems, etc; NIF staff will consider this in planning evaluation**

Summary Shot Table	<i>FY2010</i>	<i>FY2011</i>	<i>FY2012</i>	Comments
Total shots	<i>0</i>	<i>5</i>	<i>5</i>	

Summary of proposed experiment (Page 2 of 3)



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- Brief campaign description (include summary of preparatory shots (drive, diagnostic development, other) and actual data acquisition shots):

- 2010—We will converge on a diagnostic that will make it through the TALIS review. We have 3 conceptual designs and have begun to evaluate the feasibility of each.

- 2011—We will develop and impliment the chosen diagnostic and begin to take diffraction data. We will demonstrate that the signal and background levels are sufficient for quantitative liquid x-ray diffraction. Afterwards we will attempt to drive and measure diffraction through the proposed liquid-liquid transition in MgSiO_3

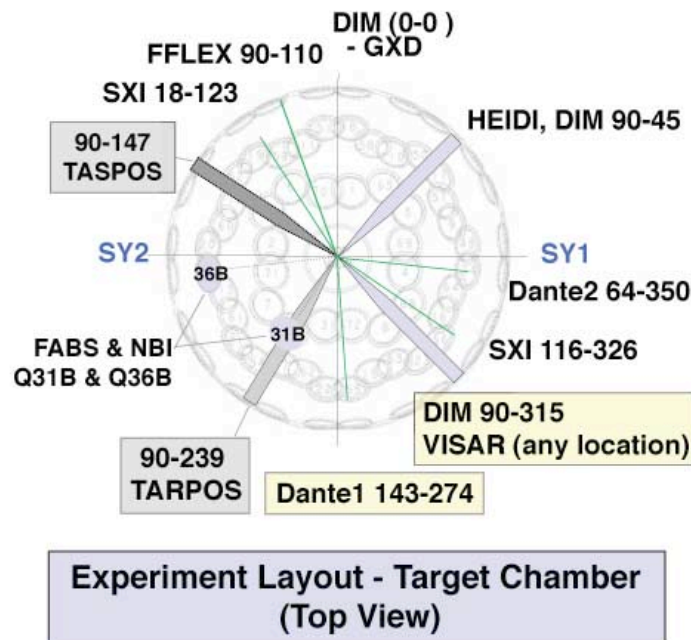
- 2012—We will continue to explore the liquid structures of MgSiO_3 . If possible we will attempt to determine the volume discontinuity by diffraction.

- We expect to be able to achieve these goals with 5 shots per fiscal year.

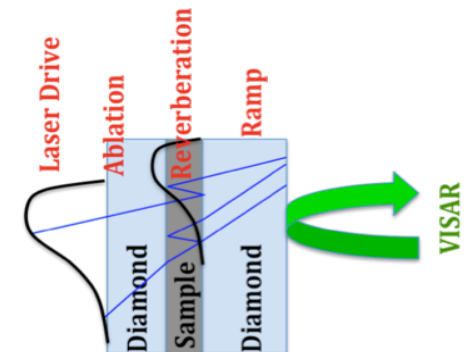
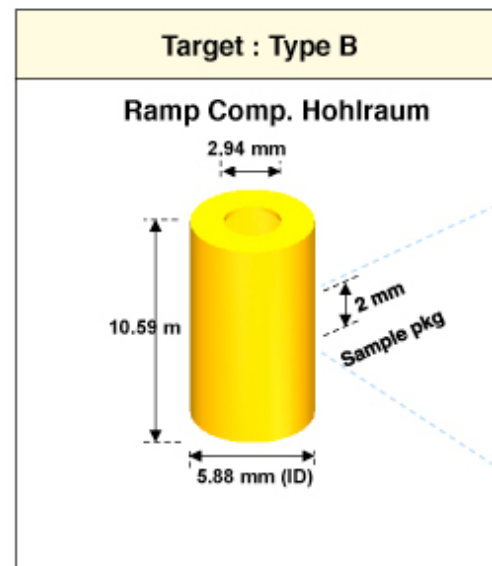
Summary of proposed experiment (Page 3 of 3)

- **Sketch of experimental configuration:** Pls. provide a simple sketch of the experimental configuration below. Include orientation of target, laser and any backlighter beams, diagnostic sightlines, etc. If configuration is identical to an existing platform so indicate. For further information on existing platforms and chamber geometry see the NIF website:

https://lasers.llnl.gov/for_users/experimental_capabilities/index.php



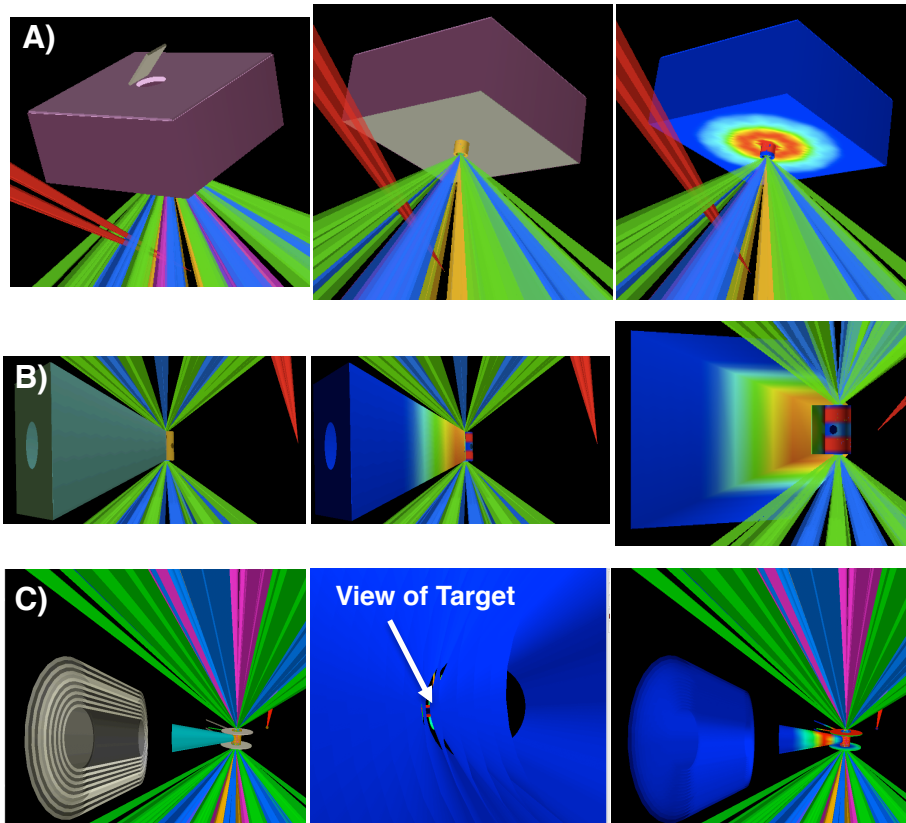
Experimental set-up: One for each unique illumination AND diag config,
e.g. if you change either, requires a different setup
Priority: (1=must have, 2=like to have, 3=ride-along)
Type: (1=New diag, 2=major mod, 3=minor mod or existing)



Possible diagnostic configurations



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Three possible configurations for an x-ray diffraction diagnostic on the NIF. A) IP box with hemispherical symmetry. Requires a 90° VISAR mirror, on-axis x-ray source, and detailed TALIS review of unconverted light, target alignment, and IP protection. B) IP pyramid, using the NIF's Hohlraum geometry, including an off-axis x-ray source. Requires a detailed TALIS review of unconverted light, target alignment, and IP protection. C) Single IP shielded by conical Soller slits, using the NIF's Hohlraum geometry, including an off-axis x-ray source, and avoiding all unconverted light. Requires a detailed TALIS review of diagnostic (Soller-slit) alignment.

Diagnostic requirements



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- Please refer to the diagnostic list on NIF user website:
https://lasers.llnl.gov/for_users/experimental_capabilities/diagnostics.php
- List below NIF diagnostics required for your experiment (along with a short summary description of required spatial, temporal, and spectral resolution) or describe what you wish to observe, and NIF staff will match to available diagnostics.

- VISAR/SOP
- DANTE
- SXI
- FABs/NBI
- FFLEX

- Also indicate below if any additional, user provided diagnostics are required. Provide a short summary of the user provided diagnostic below, including a list of all materials to be introduced into the target chamber.

- NIF XRD—Xray Diffraction Diagnostic to be developed. This diagnostic will introduce Tantalum, Stainless Steel, and Image Plates into the target chamber. We will develop a diagnostic consistent with NIF requirements

Laser requirements (1 of 2)



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Laser Parameter	Value
1) Platform to be used	<i>Materials XRD</i>
2) Number of beams required	<i>100 or 192</i>
3) 3w energy desired per beam (Maximum allowed: 3 kJ (2nsec square); for pulses other than 2nsec square provide plot of desired power vs. time on next page. NIF User Office will inform users if energy requirements exceed allowable.)	<i>0.3 to 2 kJ per beam in a 15 ns pulse consistent with the Materials EOS drive</i>
4) Peak power per beam (350 TW maximum total peak power for shaped, ignition-like pulses)	<i>1.0 TW</i>
5) Pulse shape (up to 20 nsec duration) (Options: Square, impulse (88 psec), or shaped; provide plot of desired power vs. time for shaped pulse on next page)	<i>15 ns ramped pulse shown on next page.</i>
6) SSD bandwidth (options- 45 to 90 GHz, 45 GHz default)	<i>45 GHz (modify if desired)</i>
7) Focal spot size (~250-mm (unconditioned) or ~1-mm (conditioned))	<i>1-mm</i>
9) Delays between beams (up to 10 nsec-all pulses in a quad must have same delay)	<i>Backlighter quad (35T) delayed by up to 20 ns</i>
10) Backlighter beam energy, pulse duration	<i>~1kJ, 1ns</i>
11) Other specifications	



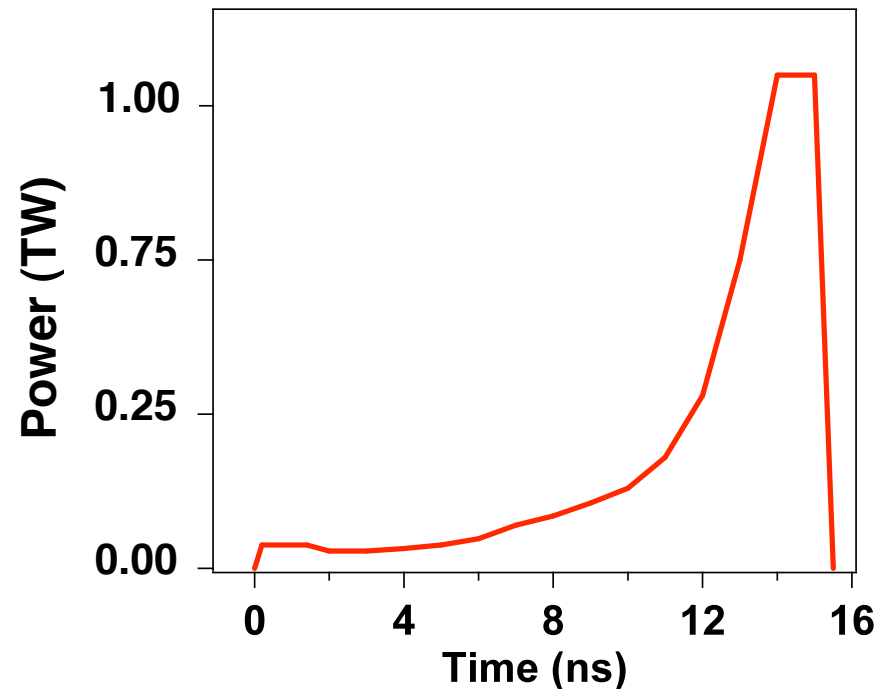
Laser requirements (2 of 2)



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For shaped pulses, sketch desired power vs. time below:

This is the maximum pulse per beam required. Shots will be range from about 10 to 100% of this.



Target requirements (1 page per target type)



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- List target types required (example: drive measurement; diagnostic test; data acquisition target)

We will use both diamond and LiF final windows for our targets. The package is 3 mm diameter, and the tantalum pinhole is 5 mm diameter.

- For each target type provide a sketch of the target below. Include dimensions and a list of *all* materials to be used. Also indicate any critical tolerances required, and indicate components (if any) to be provided by the Principal Investigator.

